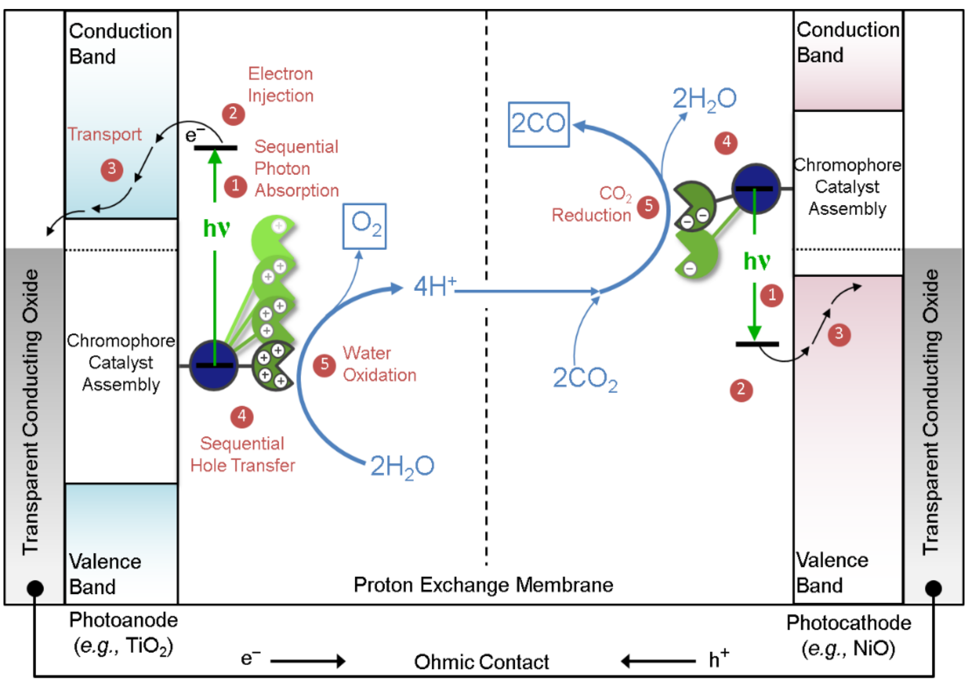
Center for Solar Fuels (UNC EFRC)
EFRC Director: Thomas J. Meyer
Lead Institution: University of North Carolina at Chapel Hill
Start Date: August 2009

Mission Statement: *To conduct research on dye sensitized photoelectrosynthesis cells (DSPECs) for water splitting and tandem cells for the reduction of carbon dioxide to carbon-based solar fuels.*

The UNC EFRC Center for Solar Fuels is conducting research on solar energy conversion and storage based on solar fuels. Research in the Center extends from the basic science of fundamental processes and new materials to integrating components into surface-stabilized assemblies, and assemblies into device prototypes. A broad, multidisciplinary approach is used in a highly collaborative setting. EFRC research integrates capabilities and expertise in chemistry, physics, and materials science with a focus on the Dye Sensitized Photoelectrosynthesis Cell (DSPEC) for solar energy conversion by water oxidation to oxygen and reduction of carbon dioxide to carbon-based fuels.



TARGET METRICS

- Converting solar photons into redox equivalents in the attached chromophore-catalyst assembly with >90% efficiency (steps **1** and **2**).
- Building up multiple redox equivalents at catalyst sites for water oxidation or CO_2 reduction with efficiencies >95% on the timescale of the light source, seconds for ambient sunlight (steps **3** and **4**).
- Achieving catalytic rates of $>10^2\ s^{-1}$ (step **5**).
- Achieving overall device efficiencies of 15% with 10^7 cycles/year and a 95% retention in photoreactivity.

Dye-Sensitized Photoelectrosynthesis Cell (DSPEC) for CO_2 Reduction to CO

THE DYE SENSITIZED PHOTOELECTROSYNTHESIS CELL (DSPEC).

The dye sensitized photosynthesis cell provides a systematic approach to solar fuels by integrating molecular light absorption and catalysis with the high bandgap properties of nanoparticle films of semiconductor oxides. Research on DSPEC device prototypes in the UNC Center for Solar Fuels utilizes an integrated, modular approach. Parallel research efforts are pursued on catalysts for water oxidation and CO_2 reduction, light-harvesting chromophores and chromophore arrays, synthesis and characterization of chromophore-catalyst assemblies, nanoparticle oxide films, core/shell structures and stabilizing overlayers by atomic layer deposition (ALD), dynamics of interfacial electron transfer, photoanodes for water oxidation, photocathodes for CO_2 reduction, and assembly and evaluation of DSPEC device prototypes.

Current research builds on the results of continued catalyst development for CO_2 reduction and water oxidation, targeting catalysts that are stable through 100,000s of turnovers at rates that exceed 10 s^{-1}. Catalysts are being integrated with organic and metal complex chromophores that are designed to absorb light broadly in the visible spectrum giving excited states capable of undergoing electron or hole injection to oxide anodes or cathodes. Assembly structures are being explored that link multiple chromophores to a single catalyst in molecular and polymer scaffolds. The successful core/shell photoanode architecture is being extended to new materials and to new core/shell structures in order to maximize solar efficiencies, and to *p*-type oxides for photocathode and tandem cell applications with a special focus on CO_2 reduction. ALD, electropolymerization, and polymer overlayer strategies are being exploited to stabilize surface structures and control interfacial properties and dynamics. The results of steady-state and time-resolved photocurrent and transient absorption measurements are being used to evaluate interfacial electron transfer dynamics and how they dictate cell performance, and to identify and overcome deleterious pathways leading to assembly instability. Measurements on device prototypes are being extended to long-term performance and to zero-gap configurations appropriate for device applications.

Research in the Center for Solar Fuels is multidisciplinary and highly integrated. The challenges posed by the research are met by the team-based structure below. Based on past success and promise for the future, the Center is poised to have, what could be, a transformative impact on solar energy conversion and storage and a role to play in the world's energy future.

RESEARCH	TEAM	RESEARCH FOCUS
Synthesis and Catalysis	Catalysis	Catalyst development and mechanistic studies on solution and interfacial catalysts for water oxidation and CO_2 reduction. Evaluation of catalysts in assemblies and device prototypes for photoanode and photocathode applications.
	Assemblies	Design, synthesis, and characterization of molecular, oligomer and polymer chromophore-catalyst assemblies for applications in water oxidation and CO_2 reduction at *n*- and *p*-type semiconductors.
Interfaces and Devices	Interface Dynamics	Provide detailed understanding of surface mechanisms that guide design of molecular systems and materials to improve functional performance of DSPEC photoanodes and photocathodes.
	Photocathode	Design, synthesis and characterization of hole-transporting semiconductor nanomaterials, core/shell structures, and light-absorbing sensitizers for high-performance photocathode applications integrated with molecular catalysts for CO_2 reduction.
	Photoanode	Optimization of solar-driven water oxidation at dye-sensitized photoanodes.

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